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Final Report for Nuclear Resonance Fluorescence Measurements of 239Pu above 2.5 MeV

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Final Report for Nuclear Resonance Fluorescence Measurements of 239Pu above 2.5 MeV

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Abstract

Nuclear Resonance Fluorescence measurements were performed at the free electron laser facility at UC Santa Barbara using a bremsstrahlung beam. Three endpoint energies were chosen for the bremsstrahlung to cover as much area above 2.5 MeV as possible. We were able to set an upper limit of NRF state strengths between 2.5 and 3.8 MeV at roughly 38(5) eV barns at the 4-sigma level and 9(2) eV barns at the 1-sigma level. Published results on states near 2.4 MeV indicate strengths about 10(2) eV barns (See Bertozzi-2). Details of the results are presented in this report. This was a collaborative effort by LLNL, Passport Systems Inc., and UC Berkeley.

Introduction

Homeland Security and National Security programs are developing systems that use nuclear resonance fluorescence (NRF) to isotopically map containers (Bertozzi, Pruet). NRF is a process in which a photon, with energy equivalent to a resonance level of a particular nucleus, excites the nucleus to the resonant level. The nucleus then decays to the ground state by a direct transition or multi-step transitions through intermediate levels. NRF is a superb signature because each nucleus has its own NRF fingerprint. Therefore, any technology that uses NRF will be isotopically sensitive. Systems that use NRF can be used for areas such as commercial certification, treaty verification, non-destructive waste and fuel assay, and certifying cargo as free of special nuclear materials (SNM).

The efficacy of NRF-based detection and assay systems depend on knowledge of NRF states in the isotope being assayed. These systems perform best when the resonance properties are matched to object being assayed or scanned. For example, resonances with very large interaction cross sections are only useful for assaying small objects or the outer skin of an object, whereas resonances with lower interaction cross sections are best suited for assaying larger objects. Also, higher energy resonances are typically better at penetrating large amounts of intervening material and there are typically fewer environmental backgrounds at higher energies. A broad research goal is to generate a library of energies and strengths so that NRF scanning systems can be optimized to the object at hand. Also, these reference materials allow enable the study of system performance for different threat spaces or assay scenarios given the NRF strengths of materials of interest and measured backgrounds. Without knowledge of NRF strengths, it cannot be known, *a priori*, what threat space NRF technologies can cover. This includes threat spaces not covered by other technologies, such as passive detection, neutron interrogation, etc. Understanding the threat space covered by NRF strengths and scanning methods will allow operators to choose a suitable tune to adequately pass or fail items of interest. A larger library to span the threat space, gives an operator a degree of freedom that other interrogation systems may have.

Results of previous measurements of NRF states in 239Pu were recently published (see Bertozzi-2), which found strong resonances with energies from 2.0-2.5 MeV. Investigations at higher energies weren't possible because the accelerator could not achieve higher energies. For many applications, the dominant background for observing resonances in the 2.0-2.5 MeV region is the radioactive decay line at 2.615 MeV present in the natural thorium decay chain. Therefore, NRF states above 2.615 MeV potentially are a better choice for maximizing the signal to noise ratio even if they are somewhat weaker than the known states below 2.615 MeV. We have performed measurements on 239Pu above 2.5 MeV at UCSB. Details of the setup and results of those measurements are given below.

Experimental Setup

The NRF measurements of 239Pu were performed at UCSB's FEL facility. A schematic of the setup is given in figure 1. Bremsstrahlung photons were used to excite NRF states in 239Pu. The electron beam was varied between 4.0 MeV and 5.3 MeV to find the energy with the best signal to noise ratio. Backgrounds were expected to be a problem given the fission thresholds and neutron separation energy at 5.5 MeV. Given the flat backgrounds (discussed below) and the energy dependence of the gamma ray detectors, the region best suited for analysis was 4.0 MeV. This gave us the best sensitivity because the efficiency of the gamma ray detectors improves at low energies. With a flat background this meant that the highest efficiency would be for lower endpoint energies.

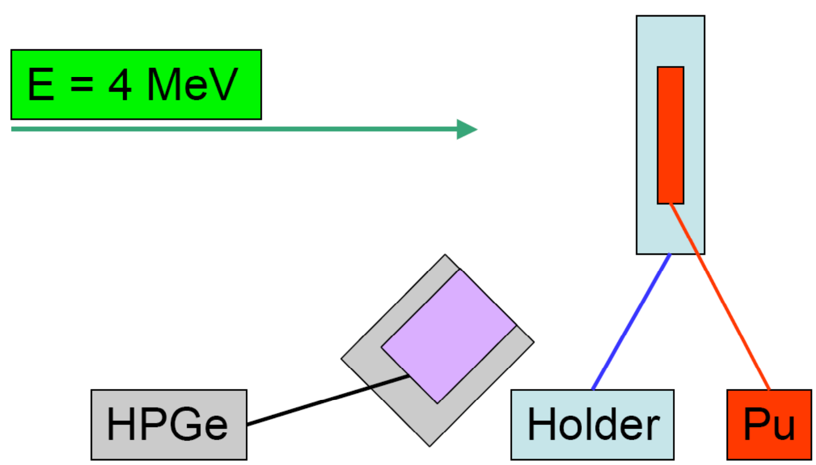

Figure 1: Schematic of the experimental setup at UCSB. Not drawn to scale.

UCSB and Passport Systems Inc. furnished the majority of the setup. The NRF target was provided by LLNL and was approximately 3.0 grams of weapons grade Pu (i.e. ~93% 239Pu). The Pu material was encased in 25.1 grams of stainless steel (Nitronic-40) in a lollipop geometry and had a thin window of 10 mils of stainless steel foil on one side, which was orientated to face the bremsstrahlung source. An aluminum disk of equal size diameter and thickness to the Pu target was placed on front of the window to normalize observed NRF peaks to the NRF state at 2211 keV in aluminum.

Gamma rays were measured from the NRF target and from background (target holder only) with a cluster of six HPGe detectors located at 120-degress with respect to the beam axis. The data acquisition system was an ORTEC single channel digital spectrometer.

Budget:

The breakdown of costs for this project is given in the table below. The total budget was $350k. The costs to date are $334k. The largest expenditure was the procurement of a subcontract with Passport Systems Inc. To date, the subcontract with Passport is $145k, which was realized in September 2008. No further collaboration with Passport is necessary for this task and the NA-22 portfolio should absorb the remainder of the lien.

Figure 2 is a graphical representation of the table below. Clearly, the largest period of spending was for the measurements in the August 2008 timeframe.

We collaborated with students and faculty members at UC Berkeley; however, they were independently funded for their efforts.

	Oct	Nov	Dec	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	YTD
ffort Costs	0	0	406	1624	4827	5733	3955	3955	368	11323	20449	11598	2873	1796	1742	7064
ravel	0	0	0	0	0	0	0	0	2198	203	2968	5148	0	248	0	1076
rocurement & Expenses	0	0	0	0	0	529	0	0	0	0	0	138441	94	5428	0	14449
acility Charges	0	0	67	273	1120	1168	712	683	-64	1577	3166	1771	61	0	0	1053
ist Direct Support	0	0	78	311	760	1273	765	761	846	2271	4430	3037	0	0	0	1453
ther Distributed	0	0	336	1349	4098	5061	3332	3312	2054	9430	19023	25596	3857	2850	2285	8258
otal Cost	0	0	887	3557	10806	13765	8764	8711	5402	24803	50035	185590	6884	10321	4027	33355
umulative Cost	0	0	887	4444	15250	29015	37779	46490	51892	76696	126731	312321	319205	329526	333553	33355
umulative Budget	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	
iens	0	0	0	0	0	0	0	147778	147778	150932	150932	112	20000	14575	15843	

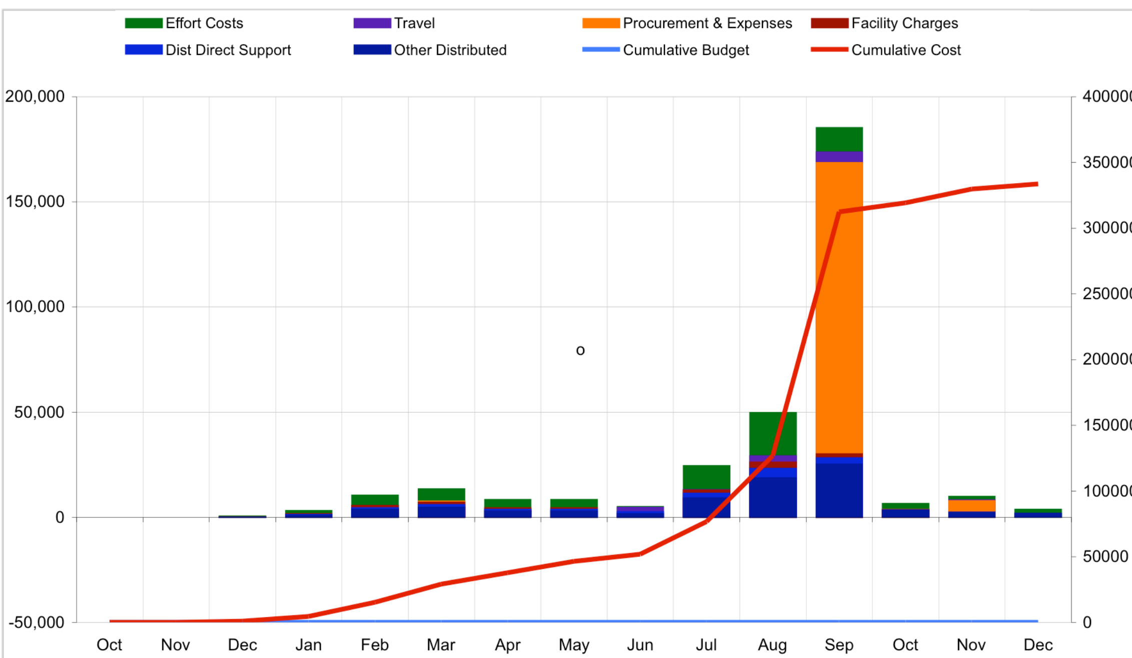

Figure 2: Breakdown of costs. Cumulative costs are depicted by the red curve. The actual numbers are given in the above table. See text for more details.

Results:

The data collected was analyzed for known and unknown peaks and structures. The region of maximum sensitivity for observing NRF scattering is between 2.4 and 3.8 MeV. The lack of photons above 3.9 MeV and the large backgrounds below 2.4 MeV rapidly reduce the sensitivity outside this window. Before undertaking the experiments, our expectation was that we would be able to observe lines with a strength greater than 8 eV barns in our measurements. However, the data collected had a high-energy background with a shape that is consistent with Compton scattering from neutron capture and inelastic processes on light nuclei (See Fig 3). Regardless of the choice for the end point energy, the Compton rate from neutron processes was not very sensitive to the end point energy. After some investigations, it was determined that the source of neutrons resulting in this high-energy background was from water in the bremsstrahlung converter. Bremsstrahung photons were causing photodisintegration of the natural abundance of deuterium in the water leading to neutron production. This background wasn't present in the previous measurements because a different design for the bremsstrahlung converter was used.

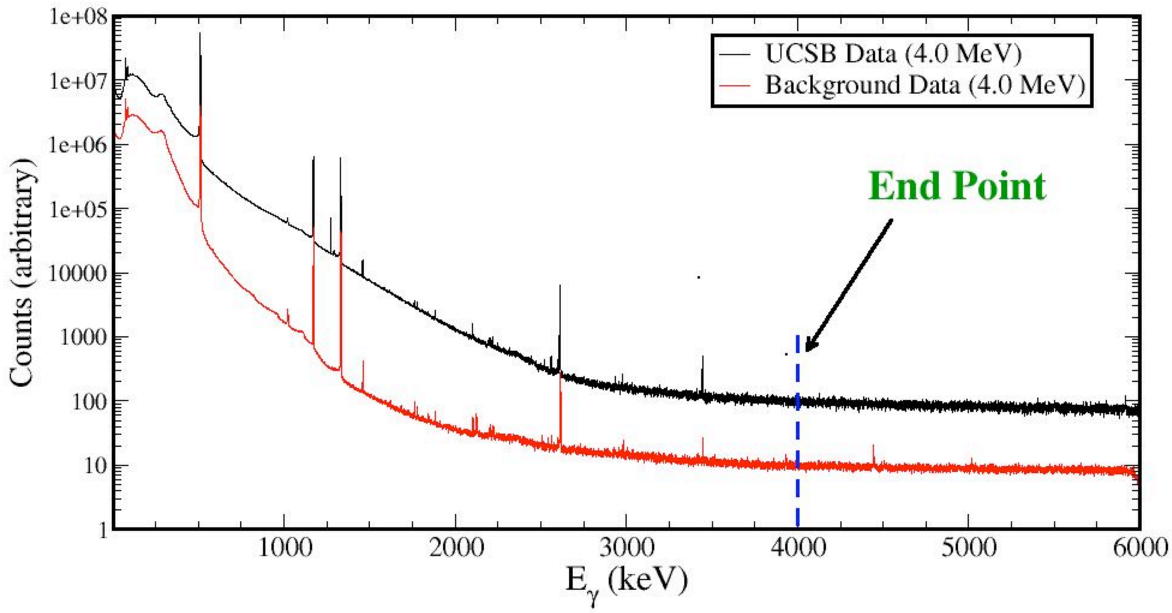

Figure 3: Comparison of UCSB data run (black) and background (red) run. The flat high-energy continuum is Compton background form high-energy gamma rays from neutron processes on light nuclei. See text for more details.

To address this large background, we focused at a low endpoint energy of 4.0 MeV where the sensitivity is the greatest. Data was collected at 4.0 MeV, and an effort was made to gain statistics and gather background data at a later date. A fraction of the background data, with the same settings as the above measurements, was collected. The background measurements used an empty holder similar to the one used for the Pu target. Figure 3 shows that the neutron background is still present.

The marked feature in both histograms in figure 3 is that there is no drop-off one would expect near the end point energy (4.0 MeV). This shows the dominance of the neutron background rate and the limitations posed to the data analysis. After a careful analysis, it was determined that additional background runs would not change the overall upper estimate on NRF states above 2.5 MeV.

However, we compared this data to our previous measurements of 239Pu (see Berotzzi-2) and the comparison is shown in figure 4. We had a sensitivity of ~18(5) eV barns for observing the previously known states at 2143 keV and 2423 keV (see Bertozzi-2) at the 1-sigma level. The strengths of these states are 13(2) eV barns and 10(2) eV barns, respectively. This is the limit at which we could have seen these peaks with the given backgrounds. The structures circled are consistent with a 1-sigma observation of those previously known lines.

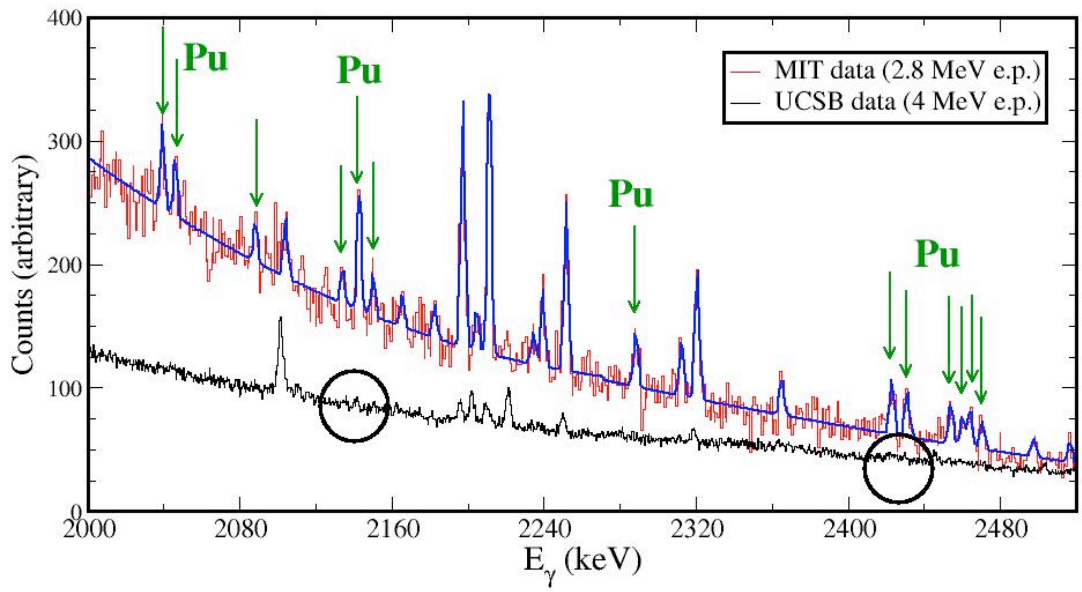

Figure 4:Comparison of Pu data from previous measurement with end point energy of 2.8 MeV (upper histogram), and this work at 4.0 MeV (lower histogram). Circled are low statistics (1 sigma) structures previously assigned to 239Pu (see Bertozzi-2).

From this data we were not able to identify NRF lines greater than 4 sigma above the previously measured lines, but we were able to set an upper limit to NRF states in the region of sensitivity (See Fig. 5). The curve was generated using the assumption that NRF "peaks" in the region of sensitivity are consistent with 1 sigma statistics. The upper limit was referenced to the observed 2211 keV NRF line in Al in a similar prescription outlined in Ref. Bertozzi-2. The energy dependent input variables used for this estimate are, detector efficiency, electronic attenuation, nuclear attenuation, flux, and Doppler broadening. The rapid increase in the cross-section estimate at 3.9 MeV is due to the cutoff at the end point energy 4.0 MeV. The uncertainties are purely statistical based on the background and the uncertainty in the 2211 keV peak area. Figure 5 indicates the nominal cross-section maximum is about 9(2) eV-barns. This estimate is consistent with average NRF states observed in Pu (see Bertozzi-2) but at a higher energy. However, for new peaks the nominal cross-section would have to be 38(5) eV barns, consistent with 4 sigma statistics, which is necessary for an acceptable confidence level.

It is also evident from figure 5 our range of sensitivity. Below 2615 keV the sensitivity curve increases as a result of backgrounds, mentioned above, and Compton continuum from the background line at 2615 keV.

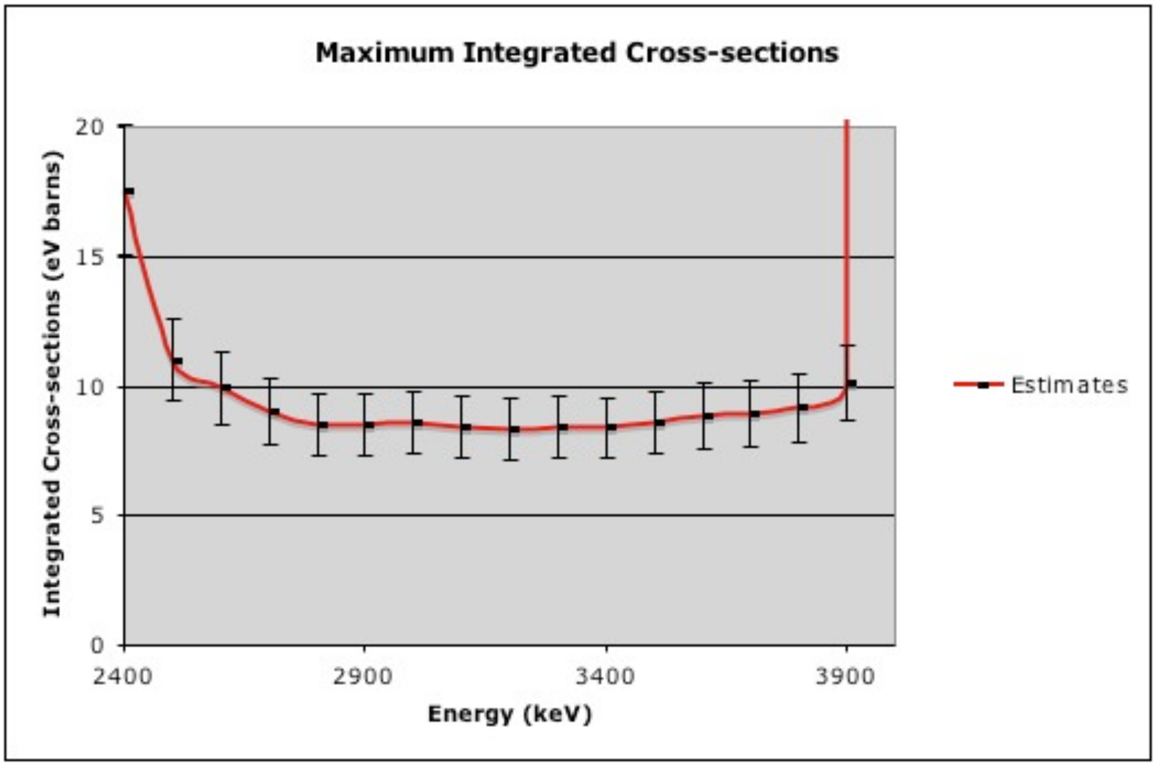

Figure 5: Estimates for maximum integrated cross sections in this region. See text for more detail.

Summary:

We have performed measurements of 239Pu with a bremsstrahlung source at UCSB to search for NRF states. Due to an unexpected neutron background, we were unable to observe any new NRF peaks between the energies of 2.5 MeV and 4.0 MeV above 4 sigma. However, we were able to set an upper limit of NRF state strengths in this energy region at roughly 38(5) eV barns. This upper bound is consistent with known neighboring NRF signatures in 239Pu (see Bertozzi-2).

James MacFarland a graduate student at UC Berkeley is carrying out further investigation of the data. His analysis will be the subject of his thesis to be submitted around May 2009.

A re-measurement of 239Pu is in order because of the unexpected background. It is important to find NRF states above 2.6 MeV to avoid natural radioactive background contamination of the signal. Furthermore, national labs should play a lead role for these efforts to accurately identify the full threat space.

References:

- W. Bertozzi and R. J. Ledoux, Nuc. Ins. Meth. Phys. Res. B 241, 820 (2005)
- Pruet *et al*. J. Appl. Phys. **99** 12310 (2006)
- W. Bertozzi(2) *et al*. Phys Rev C 78, 041601 (2008)